

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: Hessel et. al. Art Unit : 1764

Application No. : 09/898,844 Examiner : Ellen M. McAvoy

Filed : July 3, 2001

For: **COMPOSITIONS OF GROUP II AND/OR GROUP III BASE
OILS AND ALKYLATED FUSED AND/OR POLYFUSED
AROMATIC COMPOUNDS.**

DECLARATION OF Dr. EDWARD T. HESSELL UNDER 37 C.F.R. § 1.132

Commissioner of Patents and Trademarks
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Alexandria, VA 22313-1450

Mail Stop: Amendment

I, Edward T. Hessel declare and state as follows:

1. I received a Bachelor of Science degree in Chemistry in 1984 from the University of Connecticut in Storrs, Connecticut. From 1984 to 1987 I worked as a research chemist for Ciba Geigy Corp. Thereafter, I was awarded a Ph.D. in Chemistry in 1991 from the University of Rochester in Rochester, New York. Upon receiving my doctorate, I was employed for three years as a Senior Chemist for Texaco Inc. specializing in research and development for additives in motor oils.
2. From 1994 to date, I have been employed by King Industries Specialty Chemicals in Norwalk, Connecticut. I was a Senior Research Chemist, Research and Development Manager for Lubricant Additives, and was promoted

to Manager for Specialty Markets in the Research and Development Division in 2002.

3. I have read and understand U.S. Patent Application 09/898,844, of which I am a co-inventor, and the Office Action dated May 16, 2007. I understand from the May 16, 2007 Office Action that the Examiner is relying on U.S. Patent No. 6,333,298 to Waddoups et al. ("the Waddoups patent") for teaching a "lubricating oil composition" that includes a Group III base oil.
4. I have read and understand the Waddoups patent and note that at column 2, lines 45-52 patentees expressly state that the "present invention" requires at least one calcium detergent:

"The present invention **requires** the presence of at least one calcium detergent. Detergents aid in reducing deposits that build up in an engine and act as an acid neutralizer or rust inhibitor. This in turn reduces engine wear and corrosion. The use of a calcium detergent in combination with the base stock oils in the composition of this invention offers fuel economy advantages as demonstrated by coefficient of friction data."
5. I have read and understand that the Waddoups patent prefers to use a calcium sulfonate as the required detergent (column 2, lines 55-56), typically in amounts of "from 0.5 to about 5 wt. %" based on the total weight of the composition (column 3, line 26-33).
6. Based on the results of the comparative test described infra, addition of a calcium sulfonate detergent to the compositions representative of the claimed invention would adversely affect the water separation characteristics of the those compositions.

**I. WADDOUNS REQUIRED CALCIUM DETERGENT
ADVERSELY AFFECTS THE WATER SEPARATION
PROPERTIES OF THE LUBRICANT COMPOSITION**

Preparation of the Compositions Under my direction, lubricant compositions representative of those described in U.S. Patent Application 09/898,844 were prepared, i.e., Example A, Example B and Example C. The base oil combinations of a Group III base oil and alkylated naphthalene covered a range of 50-90 wt % with regard to the Group III component:

- Example A included 50 wt. % of a Group III base oil;
- Example B included 70 wt. % of a Group III base oil; and
- Example C included 90 wt. % of a Group III base oil.

In Examples A, B and C, the alkylated naphthalene is an example of a commercial embodiment of the naphthalene claimed in U.S. Patent Application 09/898,844.

Comparative examples were prepared, and are designated Examples Comp A, Comp B, and Comp C. The comparative examples Comp A, Comp B and Comp C have similar quantities of the Group III base oil and alkylated naphthalene as Examples A, B and C, but have been adjusted to contain a 400 TBN overbased calcium sulfonate, as required and described by the Waddoups patent at column 2, lines 60-63. The concentration of the overbased calcium sulfonate was adjusted to the minimum value as specified in Waddoups patent, column 3, line 32-34 (0.112% calcium). The trade name, manufacturer/supplier and physical properties of the components are shown in Tables 1-3. The quantity of each lubricant component in the lubricant composition is shown in Table 4.

TABLE 1. GROUP III BASE OIL

Trade Name: Luvodur EHVI 80

Supplier: Lehmann & Voss & Company

Property	Value
Kinematic Viscosity @ 40 °C, cSt	50
Kinematic Viscosity @ 100 °C, cSt	8
Viscosity Index	128
Pour Point, °C	-15

TABLE 2. ALKYLATED NAPHTHALENE

The alkylated naphthalene used in the lubricant compositions is an example of a commercial embodiment of the alkylated naphthalenes in U.S. Patent Application 09/898,844, Table 2, alkylated naphthalene 1 (~ 48% didodecyl naphthalene, ~ 50 polydodecyl naphthalene and < 2% monododecyl naphthalene (GC)):

Tradename: N/A

Supplier: King Industries, Inc.

Property	Value
Kinematic Viscosity @ 40 °C, cSt	100
Kinematic Viscosity @ 100 °C, cSt	11.1
Viscosity Index (calculated)	97
Pour Point, °C	-23

TABLE 3. OVERBASED CALCIUM SULFONATE DETERGENT

Trade Name: HiTEC 607

Supplier: Afton Chemical Company

Appearance	Dark brown liquid
% calcium	15.4
TBN, mg KOH/g	410
Specific Gravity, g/ml	1.21
Viscosity @ 100 °C	110

TABLE 4. LUBRICANT COMPOSITIONS

All values in weight percent

	Group III Base Oil	Alkylated Naphthalene	Overbased Calcium Sulfonate Detergent
Example A – In accordance with US 09/898,844.	50	50	0
Example Comp A	49.64	49.64	0.72
Example B - In accordance with US 09/898,844.	70	30	0
Example Comp B	69.50	29.78	0.72
Example C- In accordance with US 09/898,844.	90	10	0
Example Comp C	89.35	9.93	0.72

Evaluation of the Lubricant Compositions -Under my direction the
compositions of the invention were analyzed by ASTM Standard Test Method D 1401,

"Water Separability of Petroleum Oils and Synthetic Fluids". A copy of this test procedure is attached as Exhibit 1. This test measures the ability of petroleum oils or synthetic fluids to separate from water. Water separation is a critical aspect of many oils used in both industrial and marine applications. The lack of water separation (also termed emulsification) reduces the lubricating properties of an oil and may also cause additive instability.

A test 40 mL sample of the lubricant composition is vigorously blended with an equal volume of distilled water for 5 minutes at 54 °C in a graduated cylinder. The time required for the emulsion thus formed to separate is recorded in 5 minute increments. If complete separation or emulsion reduction to 3 mL or less does not occur after standing for 30 minutes or some other specification time limit, the volumes of oil, water, and emulsion remaining at the time are reported in milliliters (mL) in the order mL oil/mL water/mL emulsion. The complete details of this test method can be found in "Annual Book of ASTM Standards Section 5, Petroleum Products , Lubricants, and Fossil Fuels", Vol. 05.01, pages 560-563, 2007, ASTM International, West Conshohocken, PA.

**II. RESULTS OF TESTS CONDUCTED ON THE LUBRICANT
COMPOSITIONS OF TABLE 4 ARE REPORTED IN TABLE 5**

TABLE 5. ASTM D 1401 RESULTS FOR LUBRICANT COMPOSITIONS

Lubricant Composition	ASTM D 1401 Result
	Time, (Oil/Water/Emulsion)
Example A	5 minutes (41/39/0)
Example Comp A	60 minutes (10/0/70)
Example B	5 minutes (40/40/0)
Example Comp B	60 minutes (17/0/63)
Example C	5 minutes (40/40/0)
Example Comp C	60 minutes (20/0/60)

Photos of Example B and Example Comp B after 5 minutes and 60 minutes of settling are attached to further illustrate the difference in water separation properties.

Figure 1 Example B (left) and Example B Comp (right) after 5 minutes of settling in ASTM D 1401 Water Separability Test



Figure 2. Example B and Example Comp B after 60 minutes of settling in the ASTM D 1401 Water Separability Test



The results of the ASTM D 1401 test demonstrate that the lubricant compositions of U.S. Patent Application 09/898,844 show that the superior water separation properties of the claimed invention are adversely affected by the addition of a calcium sulfonate detergent, a required component of the Waddoups composition.

Date: 10/11/2007

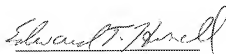

Edward T. Hessel, Ph.D.

EXHIBIT 1



Designation: D 1401 – 02

An American National Standard

Standard Test Method for Water Separability of Petroleum Oils and Synthetic Fluids¹

This standard is issued under the fixed designation D 1401; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reappraisal. A superscript epsilon (ϵ) indicates an editorial change since the last revision or reappraisal.

This standard has been approved for use by agencies of the Department of Defense.

1. Scope

1.1 This test method covers measurement of the ability of petroleum oils or synthetic fluids to separate from water.

NOTE 1—Although developed specifically for steam-turbine oils having viscosities of 28.8–90 cSt (mm²/s) at 40°C, this test method can be used to test oils of other types having various viscosities and synthetic fluids. It is recommended, however, that the test temperature be raised to 82 ± 1°C when testing products more viscous than 90 cSt (mm²/s) at 40°C. For higher viscosity oils where there is insufficient mixing of oil and water, Test Method D 2711, is recommended.

Other test temperatures such as 25°C can also be used.

When testing synthetic fluids whose relative densities are greater than that of water, the procedure is unchanged, but it should be noted that the water will probably float on the emulsion or liquid.

1.2 The values stated in SI units are to be regarded as the standard. The values given in parentheses are for information only.

1.3 *This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use. For specific precautionary statements, see 6.3–6.5.*

2. Referenced Documents

2.1 ASTM Standards:

- D 665 Test Method for Rust-Preventing Characteristics of Inhibited Mineral Oil in the Presence of Water²
- D 1141 Practice for the Preparation of Substitute Ocean Water³
- D 1193 Specification for Reagent Water⁴
- D 2711 Test Method for Demulsibility Characteristics of Lubricating Oils²
- D 4057 Practice for Manual Sampling of Petroleum and Petroleum Products⁵

¹ This test method is under the jurisdiction of ASTM Committee D02 on Petroleum Products and Lubricants and is the direct responsibility of Subcommittee D02.C0.02 on Corrosion and Water/Air Separability.

Current edition approved Nov. 10, 2002. Published December 2002. Originally approved in 1964. Last previous edition approved in 1998 as D 1401–98.

² Annual Book of ASTM Standards, Vol 05.01.

³ Annual Book of ASTM Standards, Vol 11.02.

⁴ Annual Book of ASTM Standards, Vol 11.01.

⁵ Annual Book of ASTM Standards, Vol 05.02.

3. Summary of Test Method

3.1 A test specimen consisting of a 40-mL sample and a 40-mL quantity of distilled water (Note 3) are stirred for 5 min at 54°C (Note 1) in a graduated cylinder. The time required for the separation of the emulsion thus formed is recorded either after every 5 min or at the specification time limit. If complete separation or emulsion reduction to 3 mL or less does not occur after standing for 30 min or some other specification time limit, the volumes of oil (or fluid), water, and emulsion remaining at the time are reported.

4. Significance and Use

4.1 This test method provides a guide for determining the water separation characteristics of oils subject to water contamination and turbulence. It is used for specification of new oils and monitoring of in-service oils.

5. Apparatus

5.1 *Cylinder*, 100-mL, graduated from 5 to 100 mL in 1.0-mL divisions, made of glass, heat-resistant glass,⁶ or a chemical equivalent. The inside diameter shall be no less than 27 mm and no more than 30 mm throughout its length, measured from the top to a point 6 mm from the bottom of the cylinder. The overall height of the cylinder shall be 225 to 260 mm. The graduation shall not be in error by more than 1 mL at any point on the scale.

5.2 *Heating Bath*, sufficiently large and deep to permit the immersion of at least two test cylinders in the bath liquid up to their 85-mL graduations. The bath shall be capable of being maintained at a temperature of 54 ± 1°C (Note 1), and shall be fitted with clamps which hold the cylinder in a position so that the longitudinal axis of the paddle corresponds to the vertical center line of the cylinder during the stirring operation. The clamps shall hold the cylinder securely while its contents are being stirred.

5.3 *Stirring Paddle*, made of chromium-plated or stainless steel and conforming to the following dimensions:

Length, mm (in.)	120 ± 1.5 (4 3/4 ± 3/16)
Width, mm (in.)	19 ± 0.5 (3/4 ± 3/16)
Thickness, mm (in.)	1.5 (1/16)

⁶ Borosilicate glass has been found satisfactory for this purpose.

It is mounted on a vertical shaft of similar metal, approximately 6 mm ($\frac{1}{4}$ in.) in diameter, connected to a drive mechanism which rotates the paddle on its longitudinal axis at 1500 \pm 15 rpm. The apparatus is of such design that, when the cylinder is clamped in position and the paddle assembly is lowered into the cylinder, a positive stop engages and holds the assembly when the lower edge of the paddle is 6 mm from the bottom of the cylinder. During the operation of the stirrer, the center of the bottom edge of the paddle shall not deviate more than 1 mm from the axis of rotation. When not in operation, the paddle assembly can be lifted vertically to clear the top of the graduated cylinder.

6. Reagents

6.1 *Purity of Reagents*—Reagent grade chemicals shall be used in all tests. Unless otherwise indicated, it is intended that all reagents shall conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society, where such specifications are available.⁷ Other grades may be used, provided it is first ascertained that the reagent is of sufficiently high purity to permit its use without lessening the accuracy of the determination.

6.2 *Purity of Water*—Unless otherwise indicated, reference to water shall be understood to mean reagent water as defined by Type II in Specification D 1193.

6.3 *Cleaning Solvents, Light-Hydrocarbon*, such as precipitation naphtha (**Warning**—Health hazard) for petroleum oils. Use other appropriate solvents for dissolving synthetic fluids.

6.4 *Acetone*, (**Warning**—Health hazard, Flammable.)

6.5 *Cleaning Reagent*, Cleaning by either hot NOCHROMIX⁸ (**Warning**—Corrosive. Health hazard oxidizer), or a 24-h soak at room temperature in MICRO⁹ solution gave acceptable, statistically equivalent results in round-robin testing.

7. Sampling

7.1 The test is very sensitive to small amounts of contamination. Take samples in accordance with Practice D 4057.

8. Preparation of Apparatus

8.1 Clean the graduated cylinder by removing any film of oil (or fluid) with cleaning solvent followed by a wash first with acetone and then with tap water. The glassware shall be further cleaned with a suitable cleaning reagent. Rinse thor-

oughly with tap water and then with reagent water. Inspect the cylinders for any residue or water droplets adhering to the inside walls. Both conditions indicate a need for additional cleaning.

8.2 Clean the stirring paddle and shaft with absorbent cotton or tissue wet with cleaning solvent and air dry. Care must be taken not to bend or misalign the paddle assembly during the cleaning operation.

9. Procedure

9.1 Heat the bath liquid to $54 \pm 1^\circ\text{C}$ (Note 1) and maintain it at that temperature throughout the test. Add reagent water (Note 2 and Note 3) to the graduated cylinder up to the 40-mL mark and then add to the same cylinder a representative sample of the oil (or fluid) under test until the top level of the oil reaches the 80-mL mark on the cylinder. Place the cylinder in the bath and allow the contents to reach bath temperature. Heating time may vary with type of equipment and can reach up to 30 min.

NOTE 2—If initial volumetric measurements are made at room temperature, expansion occurring at the elevated test temperature will have to be considered. For example, there will be a total volumetric expansion of about 2 to 3 mL at 82°C . Corrections to each volume reading at 82°C , therefore, should be made so that the total of the volume readings made for oils (or fluid), water, and emulsion does not exceed 80 mL. An alternative procedure which would avoid the corrections is to make the initial volumetric measurements at the test temperature.

NOTE 3—A 1 % sodium chloride (NaCl) solution or synthetic sea water, as described in Practice D 1141 or Test Method D 665, can be used in place of distilled water when testing certain oils or fuels used in marine applications.

9.2 Clamp the cylinder in place directly under the stirring paddle. Lower the paddle into the cylinder until the stop engages at the required depth. Start the stirrer and a stop watch simultaneously and adjust the stirrer, as required, to a speed of 1500 ± 15 rpm. At the end of 5 min, stop the stirrer and raise the stirring assembly until it is just clear of the graduate. Wipe the paddle with a policeman (Note 4), allowing the liquid thus removed to drop back into the cylinder. Remove the cylinder from the retaining clamps and transfer it carefully to another section of the bath. At 5-min intervals, or at the specification time limit identified for the product being tested, lift the cylinder out of the bath (see Note 5), inspect, and record the volumes of the oil (or fluid), water, and emulsion layers.

NOTE 4—The policeman should be made of material resistant to the oil or fluid.

NOTE 5—It is not necessary to lift the cylinders out of the bath for inspection if the heating bath is constructed with at least one transparent side that allows for clear visual inspection of the oil (fluid), water, and emulsion layer volumes while the cylinder remains immersed in the bath.

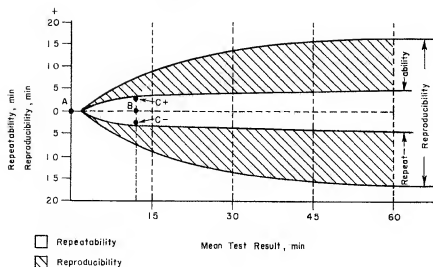
10. Report

10.1 *Recording Measurements at 5-min Intervals*—Record the time until either (1) the product passes the water separability requirements it is being tested against, or (2) the test limit for water separability is exceeded (usually 3-mL emulsion or less for 30 min at 54°C and 60 min at 82°C). The maximum volume to be reported as the oil layer is 43 mL (see Note 6). For uniformity, test results may be reported in the manner shown in the following examples:

⁷ *Reagent Chemicals, American Chemical Society Specifications*, American Chemical Society, Washington, DC. For suggestions on the testing of reagents not listed by the American Chemical Society, see *Anal. Standards for Laboratory Chemicals*, BDH Ltd., Poole, Dorset, U.K., and the *United States Pharmacopoeia and National Formulary*, U.S. Pharmacopoeial Convention, Inc. (USPC), Rockville, MD.

⁸ The sole source of supply of NOCHROMIX known to the committee at this time is Godax Laboratories Inc., 720-B Erie Ave., Takoma Park, MD 20912. If you are aware of alternative suppliers, please provide this information to ASTM International Headquarters. Your comments will receive careful consideration at a meeting of the responsible technical committee¹, which you may attend.

⁹ The sole source of supply of MICRO known to the committee at this time is International Products Corp., P.O. Box 70, Burlington, NJ 08016. If you are aware of alternative suppliers, please provide this information to ASTM International Headquarters. Your comments will receive careful consideration at a meeting of the responsible technical committee¹, which you may attend.



Use of Chart—Calculate the mean test result in minutes. Enter chart at the zero point, A, on the ordinate and move to the right on the abscissa to point B. Compute and locate the deviation points C+ and C-, from the mean test result. If the deviation points fall within the repeatability area, then the results are within the precision of the test.

Example—An oil has emulsion of 40-40-0 (10 min) and 40-40-0 (15 min). The mean test result is 12.5 min (B) and the deviation from the mean is +2.5 (C+) and -2.5 (C-).

These points fall within the repeatability area.

Use this graph similarly for the reproducibility of means of different laboratories.

FIG. 1 Chart for Determining Test Precision

40-40-0 (20)	Complete separation occurred in 20 min. More than 3 mL of emulsion had remained at 15 min.
39-38-3 (20)	Complete separation had not occurred, but the emulsion reduced to 3 mL, so the test was ended.
39-35-6 (60)	More than 3 mL of emulsion remained after 60 min—39 mL of oil, 35 mL of water, and 6 mL of emulsion.
41-37-2 (20)	Complete separation had not occurred but the emulsion layer reduced to 3 mL or less after 20 min.
43-37-0 (30)	The emulsion layer reduced to 3 mL or less after 30 min. The emulsion layer at 25 min exceeded 3 mL, for example, 0-36-44 or 43-33-4.

10.1.1 Recording Measurements at the Specification Time Requirement Only—Record the volumes for oil (or fluid), water, and emulsion layers at the specification time limit and determine whether (1) the product passes the water separability requirements it is being tested against, or (2) the test limit for water separability is exceeded (usually 3-mL emulsion or less for 30 min at 54°C and 60 min at 82°C). The maximum volume to be reported as the oil layer is 43 mL (see Note 6). For uniformity, test results may be reported in the manner shown in the examples provided in 10.1.

10.2 The appearance of each layer may be described in the following terms:

10.2.1 Oil (or Oil Rich) Layer:

10.2.1.1 Clear.

10.2.1.2 Hazy (Note 7).

10.2.1.3 Cloudy (or milky) (Note 7).

10.2.1.4 Combinations of 10.2.1.1-10.2.1.3.

10.2.2 Water or Water-Rich Layer:

10.2.2.1 Clear.

10.2.2.2 Lacy or bubbles present, or both.

10.2.2.3 Hazy (Note 7).

10.2.2.4 Cloudy (or milky) (Note 7).

10.2.2.5 Combinations of 10.2.2.1-10.2.2.4.

10.2.3 Emulsion:

10.2.3.1 Loose and lacy.

10.2.3.2 Cloudy (or milky) (Note 8).

10.2.3.3 Creamy (like mayonnaise) (Note 8).

10.2.3.4 Combinations of 10.2.3.1-10.2.3.3.

NOTE 6—Certain oils may produce a hazy oil layer. In situations where the measurement of the oil and water layer indicates essentially complete separation, the upper layer should be reported as oil. If there are two layers and if the upper layer is more than 43 mL, this layer should be considered the emulsion layer.

NOTE 7—A hazy layer is defined as being translucent and a cloudy layer opaque.

NOTE 8—The principal difference between cloudy and creamy emulsions is that the former is quite fluid and probably unstable while the latter has a thick consistency and is probably stable. A cloudy emulsion will readily flow from an inclined graduate while a creamy emulsion will not.

10.3 The appearance of the oil/emulsion and water/emulsion interfaces may be described in the following terms:

10.3.1 Well-defined, sharp.

10.3.2 Ill-defined, bubbles.

10.3.3 Ill-defined, lace.

10.4 Report the test temperature if other than 54°C and the aqueous medium if other than distilled water.

11. Precision and Bias

11.1 Precision—The precision of this test method was obtained on steam-turbine oils having viscosities of 28.8 to 90 cSt (mm²/s) at 40°C and using time to 3 mL or less emulsion as the test completion. This precision is expressed graphically in Fig. 1. The graph shows the maximum allowable deviation in minutes for repeatability and reproducibility (95 % confidence) from mean emulsion test results for these oils. It may not be applicable to other oils or fluids.



11.2 *Bias*—The procedure in this test method for measuring water separability has no bias because the value for water separability is defined only in terms of the test method.

12. Keywords

12.1 emulsion; petroleum oils; steam-turbine oils; synthetic fluids; water separability

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